



IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

#9
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In Re Application To Reissue)
U.S. Patent No. 4,912,155)
issued March 27, 1990)
Inventor: Lester P. J. Burton) Group Art Unit 122
Serial No.: 07/714,441) Examiner: D. Daus
Filed: June 13, 1991)
For: Antioxidant Aromatic)
Fluorophosphites)

Honorable Commissioner of Patents
and Trademarks
Washington, D.C. 20231

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Sir:

DECLARATION OF VINCENT J. GATTO

I, Vincent J. Gatto, hereby declare as follows:

1. I am a chemist by profession and received a Ph.D. degree in organic synthesis from the University of Maryland, completing my requirements for the degree in 1984. I worked as a post-doctoral research associate for the University of Miami from December, 1984, until February, 1987. I have been employed as a research chemist by Ethyl Corporation since February, 1987, now having the title of Research Specialist. My research has been in the field of antioxidants throughout my employment by Ethyl Corporation.

2. The following experiments were performed by me or under my supervision:

EXPERIMENT I

A 2g sample 92% 2,2'-ethylidenebis (4,6-di-t-butylphenyl)chlorophosphonite was stirred in a two-phase solvent system consisting of 15g of methylene chloride and 5g of distilled water for 20 hours at reflux temperature and then cooled. After the phases were separated, the organic phase was dried with MgSO₄ and analyzed by ³¹P-NMR for the original halophosphonite and the hydrogen phosphonate (HP) hydrolysis product. The results of the analysis are shown below.

EXPERIMENT II

Experiment I was repeated except that the compound of Experiment I was replaced with a 2g sample of 99.9% 2,2'-ethylidenebis(4,6-di-t-butylphenyl)fluorophosphonite.

3. The results of the analysis are shown below:

ANALYTICAL RESULTS AFTER 20 HOURS

Exp.	Halogen	% Unhydrolyzed Halophosphonite	% Hydrolysis Product (HP)	% Hydrolysis
I	Cl	11.0	88.9	81.0
II	F	99.8	0.2	0.1

4. As demonstrated by these experiments, the fluorophosphonite of Experiment II is much more hydrolytically stable than the chlorophosphonite of Experiment I. These experiments further demonstrate that, as a practical matter, the chlorophosphonite analog is too unstable hydrolytically to be suitable for use as a polymer antioxidant. The extensive hydrolysis

observed in Experiment I indicates that during compounding operations in the presence of any moisture, a factor which can not be reasonably avoided in the manufacture, transport or use in polymer processing plants, the chlorophosphonite will liberate hydrogen chloride. As a result the chloro-analog will degrade to hydrogen phosphonate, and will liberate HCl. The HCl will adversely affect the polymer composition and will cause costly corrosion of polymer processing equipment.

5. I believe that the above experiments are illustrative of a generic difference between the fluorophosphonites and their chloro analogs. In our work we do not take any particular precautions to avoid moisture contact with the fluorophosphonites. With chlorophosphonites, on the other hand, we have found it necessary to avoid any moisture contact, because moisture produces hydrogen phosphonates which is indicative of hydrolysis.

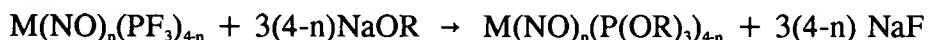
6. As part of our work related to the above experiments, I also attempted to prepare other halo-phosphonite analogs of 2,2'-ethylidenebis (4,6-di-t-butylphenyl) fluorophosphonite. Work on the preparation of the iodo derivative was not attempted because of the danger in handling PI₃. (It is a powerful deoxygenating agent.) I attempted, however, to prepare a Br analog and the following work was done in April 1991. To the best of my information, no one at Ethyl had attempted any experiments with PBr₃ before these following experiments. First, I attempted to react PBr₃ with 2,2'-ethylidenebis (4,6-di-t-butylphenol) at reflux in methylene chloride, but there was no reaction.

Second, I attempted to run the reaction in refluxing xylene with pyridine catalyst. At that point, large amounts of unreacted PBr₃, hydrogen phosphonate and complicated structures were obtained, in addition to what appeared to be the desired material according to NMR. I would not consider the reaction successful because of the low yield and the byproducts obtained.

Third, PBr₃ was added to a stirred solution of 2,2'-ethyldenebis (4,6-di-t-butyl phenol) and a stoichiometric amount of triethylamine in methylene chloride. Heat was generated and the reaction was allowed to cool to room temperature. This resulted in the formation of triarylphosphites as the major product. Again, I could not confirm the presence of the desired material.

7. This information was conveyed to Richard Glass, Dr. Kestusis Keblys and Dr. John C. Wollensak at Ethyl shortly after the completion of the experiments.

8. I have also conducted a literature survey to determine any successful reactions between PF₃ and a phenol. No such literature has been found. The closest literature was Kruck, V. et. al., Ch. Z. Anorg. Allg. Chem., 1973, 402:16-28, which indicated the following:



M = Fe, Co

R = CH₃, C₂H₅, n-C₃H₇, i-C₃H₇, n-C₄H₉, C₆H₅

9. I further declare that all statements made herein of my own knowledge are true and that all statement made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application and any patent issued thereon.

February 6, 1992
Date

Vincent J. Gatto
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